# Simultaneous optical parametric oscillation and intracavity second-harmonic generation based on a hexagonally poled lithium tantalate

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**Abstract:** Simultaneous optical parametric oscillation and intracavity second-harmonic generation based on a hexagonally poled lithium tantalate is reported. Both the optical parametric oscillation and the cascaded noncollinear second-order harmonic generation processes reach a high efficiency. A variety of possible self-doubling optical parametric oscillation processes indicate this hexagonally poled lithium tantalate has potential applications as a compact multi-wavelength light source.

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#### 1. Introduction

Optical parametric oscillation (OPO) is a unique technique for extending the wavelength to longer extents. By adding another nonlinear crystal inside or outside the OPO cavity, cascaded up-conversion can be achieved and new wavelengths can be generated simultaneously [1-13]. It is especially useful to extend the ultrafast laser's wavelength and obtain the visible femtosecond laser [7-9]. If this cascaded process can be realized in a single nonlinear crystal, the experimental setup will be greatly simplified and much more compact. The  $\chi^{(2)}$ -modulated nonlinear crystal also called quasi-phase-matching (QPM) material is a unique candidate for this type of cascaded nonlinear process. People can play mathematical tricks on the one dimensional  $\chi^{(2)}$ -modulation such as tandem periodic [12], quasi-periodic [14], dual-periodic [15] and aperiodic [13] structures etc.. In such structures, multi-process including high-order harmonics, cascaded OPO and frequency self-doubling OPO etc. have been realized [9-15]. In recent years the QPM has been extended into the 2-dimensional (2D) structure [16], the cascaded process in 2D structure has attracted much interest [17,18].

In this paper we report the experimental realization of self-doubling optical parametric oscillation (OPO) based on a hexagonally poled lithium tantalate crystal (HexPLT) in which the optical parametric down conversion (PDC) and the second harmonic generation (SHG) are accomplished simultaneously. The high-Q cavity is designed to approach a high gain OPO and a high efficiency noncollinear intracavity SHG. This experiment results indicate another way to extend wavelength such as to obtain the blue laser with very short wavelength based on a single HexPLT crystal.

#### 2. Experimental setup

The HexPLT sample in the experiment is fabricated by electric field poling technique [19]. Figure 1(a) shows its domain structure after slightly etched in acids. The near circularly inverted domains  $(-\chi^{(2)})$  with radius r) distribute hexagonally in a  $+\chi^{(2)}$  background with structure parameter  $a=9.05 \ \mu\text{m}$ . We define the reversal parameter r/a=-28% and the corresponding duty cycle  $f = 2\pi r^2/\sqrt{3}a^2$  is then -28%. The dimension of this sample is  $15 \ \text{mm}(x) \times 5 \ \text{mm}(y) \times 0.5 \ \text{mm}(z)$ . Each reciprocal lattice vector (RLV) has other equivalent 5 ones as shown in Fig. 1(b) due to the sixfold degeneration of this structure. The general phase-matching \_condition for nonlinear interaction\_in\_the QPM material is  $k_3 - k_1 - k_2 - G_{m,n} = 0$ , where  $G_{m,n} = 4\pi(\sqrt{m^2 + n^2 + m \cdot n})/\sqrt{3}a$  is the RLV of the HexPLT with lattice parameter a, and the subscripts m and n are integers, representing the order of RLV.  $\vec{k}_1$ ,  $\vec{k}_2$  and  $\vec{k}_3$  represent the interacted three waves.

The experimental setups of the involved several optical tests are rather simple. The pump is incident along  $\hat{x}$ -axis by a focus lens and the polarization direction of the pump is along the  $\hat{z}$ -axis of the HexPLT. After the HexPLT we use optical filters to eliminate the pump beam.



Fig. 1. The micrograph of the hexagonally poled lithium tantalate (a) and its reciprocal space (b).

### 3. Experimental results and discussion

We first examine the detuning characters of QPM-PDC and QPM-SHG processes in this HexPLT. For the PDC process, we apply the pump beam of 10 Hz 532 nm laser with the pulse width of 3.5 ns and linewidth of 0.15 nm. We use f= 150mm lens to focus this  $\hat{z}$ -polarized pump beam crystal getting the beam waistaround 120 µm inside the HexPLT. When the pump beam incident along the direction of  $G_{0,1}$  ( $\hat{x}$ -axis) and setting the crystal temperature during 20°C ~ 200°C, a QPM collinear parametric down conversion process happens. The momentum conservation is ensured by  $k_p - k_s - k_i - G_{0,1} = 0$ . The signal and idler wavelengths vary with the temperature. The experimental data is shown in Fig. 2(a) which consists well with the theoretical calculation. A wide spectrum from 860 nm to 1380 nm is covered when the temperature varying from 20°C to 200°C.

For the QPM-SHG process, we employ a tunable laser source (Surelite -10, Continuum) and the wavelength lies within 260 nm-2600 nm. The pump beam is still incident along the direction of  $G_{0,1}$  ( $\hat{x}$ -axis) and the HexPLT temperature is set at room temperature (20°C), multiple noncollinear QPM-SHG processes happen and the momentum conservation is ensured by  $\vec{k}_2 - 2\vec{k}_1 - \vec{G}_{m,n} = 0$ . The fundamental wavelength required for each SHG and the harmonic beam's output angles are listed in Fig. 2(b). This is consistent well with the theoretical calculation. The wavelength of the second harmonic beam lying between 400 nm-700 nm covers quasi-continuously the most of the visible spectrum. The corresponding fundamental wave is in range of 800 nm ~ 1400 nm. In Fig. 2(b), the high-order RLV like  $G_{10,-5}$  can even efficiently participate into the SHG process. This shows that the HexPLT has a quite well uniformity.



Fig. 2. (a) Temperature dependant signal and idler wavelengths which are generated from the parametric down conversion process participant by RLV  $G_{0,1}$  (b) Multiple QPM-SHG processes in the HexPLT

When comparing Figs. 2(a) and 2(b), it is not hard to find that the required fundamental wavelength for SHG participant by multiple  $G_{m,n}$  just locates around the generated spectrum from PDC participant by  $G_{0,1}$ . So by inserting this HexPLT into an optical resonant cavity, a variety of SHGs coupled with OPO will happen and this is what we called frequency self-doubling OPO. This frequency self-doubling process should have a narrow temperature bandwidth since the SHG process is critical on temperature or wavelength. The SHG process participant by  $G_{1,0}$  while the OPO process accomplished by  $G_{0,1}$  should be the most intense self-doubling OPO process since both  $G_{1,0}$  and  $G_{0,1}$  contain the maximum effective nonlinearity. The d<sub>eff</sub>=0.32d<sub>33</sub> for  $G_{1,0}$  and  $G_{0,1}$  when the duty cycle  $f=\sim 28\%$ . The temperature for this simultaneous QPM-PDC and QPM-SHG is 178.8°C. And the wavelengths for signal, idler and idler harmonics are at 873 nm, 1362 nm and 681 nm respectively. The signal and idler are generated collinear with the pump beam while the idler harmonics are of mirror symmetry about the pump beam with external angle of  $\pm 5.5^{\circ}$ . We put the HexPLT into a flat-flat cavity as shown in the inset in Fig. 3.  $\omega_p$ ,  $\omega_s$ ,  $\omega_i$  and  $\mathcal{O}_{ih}$  represent the pump, the signal, the idler and the idler harmonics, respectively. The cavity mirrors  $M_1$  and  $M_2$  have high-reflection coating for the idler 1362 nm (R=99.8%) and high-transmittance coating for the signal 873 nm (T=98.3%) and the idler harmonics 681 nm (T=92.2%). The crystal end faces have antireflection coating for the idler 1362 nm and the idler's harmonics 681 nm. The residual reflectance is 0.33% and 0.17%, respectively. The pump laser is 10k-Hz 532 nm light which has the time duration of 12.2 ns and the linewidth of 0.15 nm (DS20H-532, Princeton Industries). We use the f=300 mm lens to focus the pump beam into the HexPLT getting the beam waist to be 240  $\mu$ m. The cavity's length is 60 mm around. In Fig. 3 the output power of the signal and the idler harmonics versus the pump power are given. The signal power obeys the linear relation with the pump power while the idler harmonics follows the quadratic line. The



Fig. 3. The measured signal power versus the pump power. The inset is the schematic optical alignment of this OPO cavity.

oscillation threshold is 565 mW corresponding to a peak intensity of 28.3 MW/cm<sup>2</sup>. When pump power reaching 2.37 W, we get the signal 1.04 W and idler harmonics 380 mW after taking all the linear loss mainly by mirror's reflection into account. The efficiency of pump to signal is 44.1% and the corresponding photon number conversion efficiency reaching 72%. The slope efficiency reaches 60%. The conversion efficiency from the idler to its harmonics is about 56.9%. The final conversion efficiency from the pump to the harmonics is around 24%, which is high enough even compared with the femto-second pump case [13]. Both the OPO and SHG work in a high-gain regime.

It is worth noted that here this cascaded harmonic generation contains an effective second-order nonlinearity of  $\sqrt{2} d_{eff}=0.47d_{33}$  because  $G_{1,0}$  and its mirror symmetry can participate into SHG simultaneously. When the duty cycle f=28%, the effective nonlinearity  $d_{eff}=0.32d_{33}$  which is close to its maximum. This makes the intracavity SHG very efficient. Here the HexPLT is proved to be a compact and efficient medium for self-doubling OPO. Furthermore, without an isolator the backward SHG using reciprocal vector  $G_{-1,0}$  and its mirror symmetry in this linear flat-flat cavity occurs at the same time. So in this experiment we observed not only a pair of red spots along the pump propagating direction but also a pair of them along the opposite direction. In the above experiment the power of the idler harmonics only includes the forward two red spots while the backward two spots which have equivalent power are not reckoned in. Additional isolator for the idler or a ring cavity can be used to make SHG flowing unidirectionally.

In Fig. 4 we show the experimental study on the temperature detuning character on the frequency self-doubling OPO. The maximum SHG occurs when temperature set at 178.8°C and the temperature bandwidth (FWHM) is 2°C. There is a power dip around 178.8°C for the signal when the cascaded SHG happens efficiently. This is due to the fact that the effective SHG actually means a loss of the idler and this will make the OPO work less efficient.

Our previous work on the phase-matching condition of this HexPLT gives that there is still a variety of possible SHG cascaded PDC processes [20]. For example, when temperature set at around 189 °C, the signal can be doubled through  $G_{2,1}$  and the blue laser of 433 nm can be obtained. When setting temperature around 110 °C or 89 °C, the idler doubling at 630 nm or the signal doubling at 469 nm can be realized in which  $G_{3,-1}$  and  $G_{1,1}$  are utilized for momentum conservations, respectively. Although the effective nonlinearity for such process is not as large as  $G_{1,0}$  which is used for the idler doubling at 178.8 °C, the intracavity SHG



Fig. 4. The signal power and the idler harmonics power versus temperature.

should still possess a high conversion efficiency by proper coating of the cavity mirrors. There should still be other possible self-doubling OPO processes accomplished by other RLVs although these may have lower conversion efficiency. It is very useful to supply a quasi-continuous output of signal or idler doubling in addition to a tunable output from such a compact optical cavity. This could find new applications in medical machine and meet other multi-wave output demands.

#### 4. Conclusion

In conclusion, we examined the frequency self-doubling OPO based on a single hexagonally poled lithium tantalate. Without any complicated mathematical design, this simple single period structure makes this experiment is feasible to be realized. Both the single resonant OPO and intracavity SHG work at a high-gain regime. This cascaded noncollinear SHG is especially useful for extending wavelength especially approaching the blue laser and even the deep blue. Other possible cascaded SHG processes inside this HexPLT indicate it can serve as a new multi-wave generator.

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